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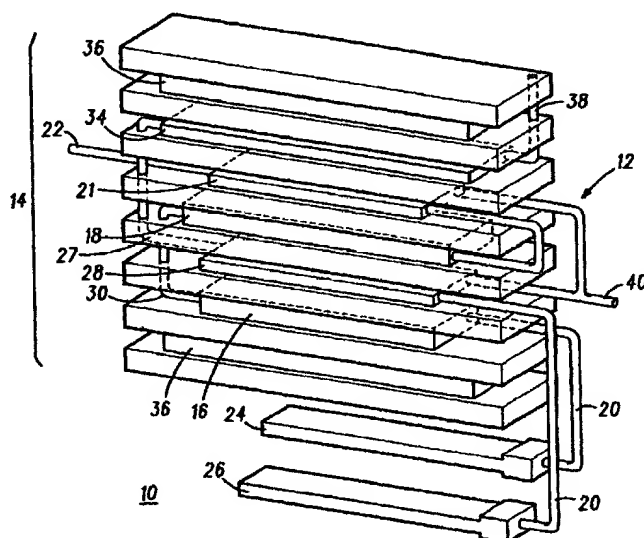
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(54) Title: **HYDROGEN GENERATOR UTILIZING CERAMIC TECHNOLOGY**



(57) Abstract: A hydrogen generator (10) including a three-dimensional multilayer ceramic carrier structure (12) defining a fuel reformer (14). The reformer includes a vaporization zone (16) and a reaction zone (18) including a catalyst. The reformer is operational as either a steam reformer, a partial oxidation reformer or an autothermal reformer. The fuel reformer, or processor, further includes an inlet channel (20) for liquid fuel and an outlet channel (22) for hydrogen enriched gas. The fuel processor is formed utilizing multi-layer ceramic technology in which thin ceramic layers are assembled then sintered to provide miniature dimensions in which the encapsulated catalyst converts or reforms inlet fuel into a hydrogen enriched gas.



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*For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.*

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HYDROGEN GENERATOR UTILIZING CERAMIC TECHNOLOGY

Field of Invention

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The present invention pertains to hydrogen generators, and more particularly to hydrogen generators utilizing multi-layer ceramic technology for improved size and performance benefits.

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Background of the Invention

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Hydrogen is attractive and has been used as the fuel in many types of devices, such as: in fuel cells for producing electricity, in combustion engines, including rockets or internal combustion engines, and in chemical processing devices, including those used in the refining of metallic components. Typically hydrogen, or more particularly, hydrogen enriched gas, is processed from methanol, natural gas, petroleum, or ammonia using a chemical reactor, typically called a hydrogen generator or reformer. A reformed hydrogen fuel cell (RHFC), utilizes hydrogen enriched gas processed from liquid or

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gaseous hydrocarbon fuels, such as methanol, using a fuel reformer.

Methanol is the preferred fuel for use in fuel reformers, or hydrogen generators, because methanol is easier to reform into hydrogen enriched gas at a relatively low temperature and yields a minimum number of gaseous by-products (carbon dioxide and carbon monoxide) compared to other hydrocarbon fuels such as natural gas, ethanol, petroleum, gasoline, or butane. This is especially important for small portable units, where the temperature of the reformer unit would be a concern. The reforming or converting of methanol into hydrogen enriched gas usually is accomplished using one of three different types of reforming processes. These three types are steam reforming, partial oxidation reforming, and autothermal reforming. Of these types, steam reforming is the preferred process for methanol reforming because it is the easiest to control, yields the minimum number of gaseous by-products (carbon dioxide and carbon monoxide), and produces a higher hydrogen output, at a lower temperature, thus lending itself to favored use. During steam reforming, raw methanol is catalytically converted, in the presence of water and with the

application of heat, to a hydrogen enriched gas.  
Although, steam reforming is the preferred process,  
partial oxidation reforming and autothermal reforming are  
utilized in many instances. During partial oxidation  
5 reforming, raw methanol is converted to a hydrogen  
enriched gas through partial oxidation of methanol over a  
catalyst in a limited supply of oxygen or air to prevent  
complete oxidation. Since this is an exothermic  
reaction, it does not require heat input to proceed. The  
10 reaction will proceed without any additional heat input  
provided the methanol and oxygen are in contact with the  
proper catalyst. Control of oxygen partial pressure and  
temperature is very critical, and for portable methanol  
reforming, the higher operating temperature of the  
15 reformer is a concern.

Autothermal reforming is a combination of the  
catalytic partial oxidation and steam reforming process.  
During the autothermal methanol reforming process, the  
partial oxidation reaction which produces heat is  
20 carefully managed to provide sufficient heat for the  
steam reforming reaction. In an autothermal reformer, the  
reactions between the input reactants, namely the  
methanol, water and air(or O<sub>2</sub>), are carefully balanced

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over the catalyst, to produce CO<sub>2</sub> and H<sub>2</sub> gases with minimum amount of CO. The partial oxidation step and steam reforming step may be done in the same or separate compartments during autothermal reforming.

5        Fuel reformers have been developed for use in conjunction with many new devices, including fuel cell devices. Many of these fuel cell devices include reformers which are typically cumbersome and complex devices consisting of several discrete sections connected  
10 together with gas plumbing and hardware to produce hydrogen gas. Accordingly, reformers have not been found suitable for portable power source applications, or in other applications requiring minimal size and weight. To date, no fuel reformer has been developed utilizing  
15 ceramic monolithic structures in which the miniaturization of the reformer has been achieved. Laminated ceramic components containing miniature channels and other features which utilize low pressure lamination ceramic technology, are now commonly being  
20 developed for use in microfluidic management systems. Monolithic structures formed of these laminated ceramic components provide for three-dimensional structures that are inert and stable to chemical reactions and capable of

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tolerating high temperatures as well as providing for miniaturized structures, with a high degree of electronic circuitry or components embedded or integrated into such a ceramic structure for system control and functionality.

5 Additionally, the ceramic materials used to form ceramic devices, which have microchannels formed within the structure, are considered to be excellent candidates for catalyst supports in microreactor devices for generating hydrogen for supplying miniaturized fuel cells.

10 Accordingly, it is an object of the present invention to provide for a miniaturized hydrogen generator, or fuel processor, that provides for the reforming of a fuel to a hydrogen enriched gas.

It is yet another object of the present invention to  
15 provide for a monolithic structure for the reforming of a fuel to a hydrogen enriched gas.

It is still another object of the present invention to provide for a monolithic structure that is formed by utilizing ceramic technology, thereby providing for the  
20 integration of a plurality of internal plumbing interconnections and electrical circuitry and connections.

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It is another object of the present invention to provide for a hydrogen generator, or fuel processor, that is miniaturized for use in conjunction with: (i) fuel cells for portable device applications; (ii) combustion  
5 devices; (iii) chemical processing devices; and (iv) other devices in which hydrogen enriched gas is consumed as fuel.

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#### Summary of the Invention

The above problems and others are at least partially solved and the above purposes and others are realized in a hydrogen generator including a three-dimensional  
15 multilayer ceramic carrier structure defining a fuel reformer, or fuel processor. The fuel reformer includes a vaporization zone and a reaction zone including a reforming catalyst. The ceramic carrier further includes an inlet channel for liquid fuel and an outlet channel  
20 for the hydrogen enriched gas. The fuel reformer is formed utilizing ceramic technology in which thin ceramic layers are assembled then sintered to provide miniature



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dimensions in which the encapsulated catalyst converts or reforms inlet fuel into a hydrogen enriched gas.

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Brief Description of the Drawings

The novel features believed characteristic of the invention are set forth in the claims. The invention itself, however, as well as other features and advantages thereof will be best understood by reference to detailed descriptions which follow, when read in conjunction with the accompanying drawings, wherein:

FIG. 1 is a simplified orthogonal view of a hydrogen generator including a plurality of channels, according to the present invention;

FIG. 2 is a simplified orthogonal view of an alternative embodiment of the hydrogen generator according to the present invention;

FIG. 3 is a simplified orthogonal view of an alternative embodiment of the hydrogen generator according to the present invention;

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FIG. 4 is a simplified sectional views of a thermally conductive channels or vias according to the present invention;

FIGS. 5 and 6 are simplified sectional views of alternative embodiments of thermally controlled vaporizer/reaction zone channels according to the present invention; and

FIG. 7 is a simplified flow diagram of a method of reforming fuel according to the present invention.

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#### Description of the Preferred Embodiments

Turning now to the drawings, and in particular FIG. 1, illustrated in simplified orthogonal view is a hydrogen generator 10 including a plurality of channels, according to the present invention. Hydrogen generator 10 is comprised of a three-dimensional multi-layer ceramic structure 12. Ceramic structure 12 is formed utilizing multi-layer laminate ceramic technology. Structure 12 is typically formed in component parts which are then sintered in such a way as to provide for a monolithic structure. Ceramic structure 12 has defined

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therein a fuel reformer, or fuel processor, generally referenced 14. Fuel reformer 14 includes a vaporization chamber, or vaporization zone, 16, a reaction chamber, or reaction zone, 18, and an integrated heat source 28.

5 Ceramic structure 12 further includes at least one fuel inlet channel 20 in communication with a liquid fuel source of a combination solution methanol/water source 24 and a pure methanol source 26 and a hydrogen enriched fuel outlet channel 22. It should be understood that in a  
10 different embodiment, separate methanol and pure water fuel sources can be used. In this instance the methanol would need to be in direct communication with the heater and a mixer would need to be utilized for mixing of the methanol and water fuel source.

15 During operation, heat is efficiently transferred from the central aspect of the device, more particularly an integrated heater 28, to the reaction zone 18 and fuel vaporizer, or vaporization zone 16 using thermal conductive channels, or vias, (discussed presently). In  
20 this particular embodiment, integrated heater 28 is described as a chemical heater, including a catalyst and arranged so as to oxidize fuel to produce heat, but it should be understood that the integration of an

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electrical heater is anticipated by this disclosure. Chemical heater 28 includes an air port 40 for providing oxygen for oxidation of fuel 24 and/or 26 and an inlet channel 20, for providing fuel 24 and/or 26 to heater 28.

5        Output from fuel vaporizer zone 16 travels via structure 30 to reaction zone 18 and then through hydrogen enriched gas outlet channel 22. Optionally included is a preferential oxidation reactor 21. During operation, output from reaction zone 18 travels to  
10        preferential oxidation reactor 21 where CO is preferentially cleaned up from the output gasses to below 30 ppm for fuel cell use. Alternatively, when a high temperature fuel cell is utilized, design 10 would not include preferential oxidation reactor 21 due to the  
15        ability for high temperature fuel cells to tolerate up to a combined total of 5% (mol) CO and methanol. Additionally, included in design 10 is a waste heat recovery zone 34 for the capture of heat from spent gases from an external device, such as a fuel cell stack, (not  
20        shown) through an inlet channel (not shown) and from chemical heater 28 through channel 27.

      An efficient thermal insulator 36 is positioned about fuel reformer 14 to keep outer temperatures low for

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packaging and also to keep heat localized to the fuel reformer. In this particular embodiment, the fuel processor operates at a temperature ranging from ambient to 300°C unless it is integrated with a high temperature fuel cell where the fuel processor operates in a range of 140-300°C. Fuel vaporizer zone 16 operates at a temperature ranging from 120-160°C and reaction zone 18 operates at a temperature ranging from 200-250°C. Additionally, in this particular embodiment of fuel processor 10, included is an exhaust gas vent 38, for the venting of exhaust gases generated by device 10.

It should be understood that alternative embodiments encompassing alternative fuel delivery means and fuel vaporizer, reaction zone, and chemical heater positions are anticipated by this disclosure. In particular, anticipated is an embodiment in which only a single fuel supply, namely methanol and water, is anticipated. This use of a single methanol and water solution would enable the fabrication of a simpler design, without any need for the device to incorporate two fuel tanks. Although it is understood that pure methanol is more efficient and preferred with respect to the chemical heater, a 1 mole water and 1 mole methanol solution will also work, but is

not deemed literally as <sup>12</sup> operationally efficient. Further, a heater using the water and methanol solution is suitable for practical applications, and would permit a simple common fuel reservoir for feeding the heater and  
5 reformer chambers. In this instance, the fuel delivery would be split into two chambers, the chemical heater 28 and fuel vaporizer 16.

Next, anticipated are variations on the actual design of device 10 and more particularly to the actual  
10 location of the fuel vaporizer zone 16, reaction zone 18 and chemical heater 28. In one particular alternative embodiment, it is anticipated that reaction zone 18 surrounds chemical heater 28 on both sides (top and bottom). In yet another alternative embodiment, it is  
15 anticipated that reaction zone 18 can be positioned below heater 28 and the fuel vaporizer zone 16 on top of the chemical heater 28. In addition, and as previously stated, it is anticipated that chemical heater 28 can alternatively be an electrical heater (not shown).

20 Finally, it is anticipated by this disclosure that although illustrated in FIG. 1 is a design in which a fuel cell is not integrated with reformer 14, it is additionally anticipated, that a fuel cell (not shown)

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can be monolithically integrated with hydrogen generator  
10. Further information on a reformed hydrogen fuel  
system device including an integrated fuel cell stack can  
be found in U.S. Patent application, bearing attorney  
5 docket number CT 00-014, simultaneously filed herewith,  
, entitled "FUEL PROCESSOR WITH INTEGRATED FUEL CELL  
UTILIZING CERAMIC TECHNOLOGY", assigned to the same  
assignee and incorporated herein by this reference.

Referring now to FIG. 2, illustrated is a partial  
10 oxidation hydrogen generator according to the present  
invention, generally referenced 50. Partial oxidation  
reformer 50 provides for part of the fuel to be oxidized  
to provide energy for the reforming reaction within the  
fuel reformer. Direct heat transfer, makes this reformer  
15 small, compact, light weight and dynamically responsive.  
Partial oxidation systems rely on the reaction of the  
feedstock in a limited supply of oxygen or air to prevent  
complete oxidation. Partial oxidation is an exothermic  
reaction and in this process, the temperature of the  
20 reformer can exceed 400°C. Control of the oxygen partial  
pressure is very critical. Partial oxidation is not  
typically used for methanol reformers, and is more  
commonly utilized in association with gasoline reformers

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for automotive applications where the temperatures can exceed 650°C.

As illustrated, hydrogen generator 50 includes a fuel 52, input through inlet 54, to a partial oxidation reactor 56. During operation, fuel 52, typically methanol, is first oxidized by a chemical heater or electrical igniter, 58 over a catalyst. Once the partial oxidation reaction starts, the heat generated will sustain the reaction. There is no need to provide additional heat as in the steam reformer described with reference to FIG. 1. During operation, the reaction needs to be controlled so that it does not proceed to complete oxidation. This is achieved by monitoring the oxygen partial pressure and temperature and by controlling the air intake 60 and fuel feed 54.

Fuel 52 travels via inlet 54 to fuel reformer 68, more particularly to first partial oxidation reactor 56 and then via channel 62, to a second partial oxidation reactor 64. An enriched hydrogen fuel outlet 66 serves to direct the reformed hydrogen fuel toward an externally connected fuel cell (not shown).

An efficient thermal insulator 70 is positioned about partial oxidation reactors 56 and 64 to keep outer



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temperatures low for packaging and also to keep heat localized to the fuel reformer. It is anticipated by this disclosure that although illustrated in FIG. 2 is a design in which a fuel cell is not integrated with reformer 68, it is additionally anticipated, that a fuel cell (not shown) can be monolithically integrated with hydrogen generator 50. In this particular design, hydrogen generator 50 includes a temperature sensor 72 and an oxygen partial pressure,  $PO_2$ , sensor 74.

10 As illustrated in FIG. 2, partial oxidation hydrogen generator 50 converts oxygen and methanol mainly into hydrogen and carbon dioxide with a small amount of carbon monoxide. Since the partial oxidation reaction is an exothermic reaction, this reforming reaction does not  
15 require the addition of heat. The reforming is performed over a temperature range of 200°C - 400°C. CO cleanup is achieved in much the same way as was done with the steam reformer illustrated in FIG. 1.

Referring now to FIG. 3, illustrated is an  
20 autothermal hydrogen generator according to the present invention, generally referenced 80. Autothermal reforming is a combination of both steam reforming, an endothermic

16  
reaction which requires heat input described with  
reference to FIG. 1, and partial oxidation reforming, an  
exothermic reaction which produces heat, described with  
reference to FIG. 2. In autothermal reforming, a balance  
5 of both of these reforming processes is achieved. More  
particularly, autothermal reforming converts fuel 82,  
more particularly water, oxygen and methanol, to hydrogen  
and carbon dioxide without the requirement of the  
addition of heat.

10 Fuel 82 travels via inlet 81 to a fuel reformer 83,  
more particularly to the first stage of the autothermal  
reactor 87 involving partial oxidation of some of the  
methanol. The fuel stream is interconnected via channel  
88, to a second stage of the autothermal reactor 90  
15 involving steam reforming of the methanol. These two  
reforming stages may occur either in the same compartment  
or in two sequential separate compartments. Either way,  
an enriched hydrogen fuel outlet 92 serves to direct the  
reformed hydrogen fuel toward an externally connected  
20 fuel cell (not shown).

Temperature sensor 84 and an oxygen partial pressure  
sensor 85 are utilized to monitor the reaction process.  
In particular, sensors 84 and 85 control the fuel feed

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(methanol and water), 82, and oxygen supply 86. If there is too much oxygen present then the reaction can proceed towards complete oxidation, consuming excess fuel, and producing unwanted excessive heat. In contrast, if  
5 sufficient heat is not produced due to partial oxidation reaction, then coking can result, hindering the steam reforming process. Autothermal methanol reforming proceeds in the temperature range of 200-300°C with the use of proper catalyst(s). CO cleanup is achieved in  
10 much the same way as was discussed with the steam reformer illustrated in FIG. 1.

Referring now to FIG. 4, illustrated in simplified cross-sectional view is a thermal transfer structure, referenced 30. Structure 30 is comprised of channels 102  
15 or vias connected with a metal film 104 in intimate contact with the various zones 18 and 28, as illustrated. It should be understood that structure 30 is generally utilized for transfer of heat efficiently between chemical heater 28 and fuel vaporizer zone 16 and  
20 reaction zone 18, as well as between waste heat recovery 34 and reaction zone 18, or any other zone where thermal transfer needs to occur. As illustrated, thermal conductive structure 30 includes multi-layer ceramic

18  
layers 100 used in the fabrication of the monolithic  
structure, referenced 12 in FIG. 1. Identified is  
chemical heater section 28 and reaction zone chamber 18.  
Thermally conductive thick-film metal vias 102 thermally  
5 couple the top and bottom sections efficiently for good  
heat transfer. A thick-film metal layer 104, in intimate  
contact with chemical heater zone 28 and reaction zone  
18, spreads and uniformly heats these sections.

Referring now to FIGs. 5 and 6, illustrated in  
10 simplified cross-sectional or plan view is a portion of  
fuel vaporizer zone 16. More particularly, illustrated  
in FIG. 5 is a serpentine channel 110, that is typically  
filled or coated with an inert porous ceramic material  
for thermal control. Alternatively, vaporizer 16 can be  
15 formed of a multiplicity of parallel channels 112, with  
an inert porous ceramic material for thermal control, in  
communication with a porous diffuser 113. In each  
instance, serpentine channel 110 or parallel channel 112  
includes a combined single liquid inlet feed 114 at one  
20 end and combining into a single vapor outlet at the other  
end 116. Serpentine channels 110 and parallel channels  
112, are formed in the multi-layer ceramic structure,  
previously identified as 12. Reaction zone 18 includes

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similarly designed channels and features as found in the vaporizer zone, discussed above. More particularly, reaction zone 18 includes a reforming catalyst. The catalyst is present as channel wall coatings or as a porous packed bed of catalyst particles. One design goal is to reduce the dimensionality of the reactor channels to the order of a gas diffusion length and control gas residence times, or gas space velocity, that are favorable to reaction kinetics. A multiplicity of parallel reactor channels, generally similar to channels 112, provide for high gas throughput and minimizes back pressure issues as related to channel blockage. It should be understood that it is anticipated by this disclosure that vaporizer zone 16 and reaction zone 18 may include in addition to, or in lieu of, serpentine channels 110 and parallel channels 112, any number of cavities or chambers, or any combination of channels, cavities, or chambers, thereof.

Illustrated in FIG. 7 in a simplified flow chart diagram, is the chemical reaction 120 that takes place in a steam reforming hydrogen generator, and more particularly in reformer 14 of device 10, as illustrated in FIG. 1. As illustrated, methanol 122 and water 124

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are input into a steam reformer 126, generally similar to reformer 14 of FIG. 1. Steam reformer 126 operates at a temperature of approximately 250°C to reform the input methanol 122 and water 124 into a reformed gas mixture, 5 generally referred to as the hydrogen enriched gas. More particularly, in the presence of a catalyst, such as copper oxide, zinc oxide, or copper zinc oxide, the methanol 122 and water 124 solution is reformed into hydrogen, carbon dioxide, and some carbon monoxide. 10 Steam reformer 126 operates in conjunction with an optional carbon monoxide cleanup 128, that in the presence of a preferential oxidation catalyst and air (or  $O_2$ ), reforms a large percentage of the present carbon monoxide into carbon dioxide. This reformed gas mixture 15 supplies fuel through fuel output 130 to an external device, such as a fuel cell.

Accordingly, described is a hydrogen generator including a ceramic carrier defining a reaction zone including a catalyst. The ceramic carrier further 20 includes a heat source thermally coupled to the reaction zone. An inlet channel is supplied for input of the liquid fuel to the fuel processor and an outlet channel

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is supplied for the output of the reformed gas mixture.  
The fuel processor device is formed as a monolithically  
integrated structure, generally comprised of a plurality  
of thin ceramic layers assembled and then sintered in  
5 such a way as to provide for the closed heating zones in  
which the encapsulated catalysts reforms the inlet fuel  
into mostly hydrogen gas suitable for supplying fuel via  
a suitable conduit to an other external device in fluidic  
communication or integrated monolithically with internal  
10 conduits.

While we have shown and described specific  
embodiments of the present invention, further  
modifications and improvements will occur to those  
skilled in the art. We desire it to be understood,  
15 therefore, that this invention is not limited to the  
particular forms shown and we intend in the appended  
claims to cover all modifications that do not depart from  
the spirit and scope of this invention.

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What is claimed is:

1. A hydrogen generator comprising:  
a ceramic carrier defining a fuel processor, the  
fuel processor including a reaction zone including a  
5 reforming catalyst;  
an inlet channel for liquid fuel; and  
an outlet channel for hydrogen enriched gas.
2. A hydrogen generator as claimed in claim 1  
10 wherein the fuel processor further includes a  
vaporization zone.
3. A hydrogen generator as claimed in claim 1  
further including an integrated heat source.  
15
4. A hydrogen generator as claimed in claim 1  
wherein the integrated heat source is a resistive heater  
that is electrically driven.
- 20 5. A hydrogen generator as claimed in claim 3  
wherein the integrated heater is a chemical heater  
including a catalyst and arranged to oxidize fuel to  
produce heat.



6. A hydrogen generator as claimed in claim 5 wherein the chemical heater further includes an air inlet for providing oxygen for the oxidation of the fuel and the inlet channel includes an opening to provide fuel to the chemical heater.

7. A hydrogen generator as claimed in claim 3 wherein the integrated heater couples heat to the reaction zone using thermally conductive structures.

8. A hydrogen generator as claimed in claim 2 wherein one of the vaporization zone and the reaction zone include a plurality of parallel channels.

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9. A hydrogen generator as claimed in claim 2 wherein one of the vaporization zone and the reaction zone include a at least one serpentine channel.

20

10. A hydrogen generator as claimed in claim 1 wherein the ceramic carrier is an monolithic three-dimensional multilayer ceramic structure.

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11. A hydrogen generator comprising:

a three-dimensional multilayer ceramic carrier structure defining a fuel reformer including a vaporization zone and a reaction zone including a  
5 reforming catalyst;

an inlet channel for liquid fuel; and

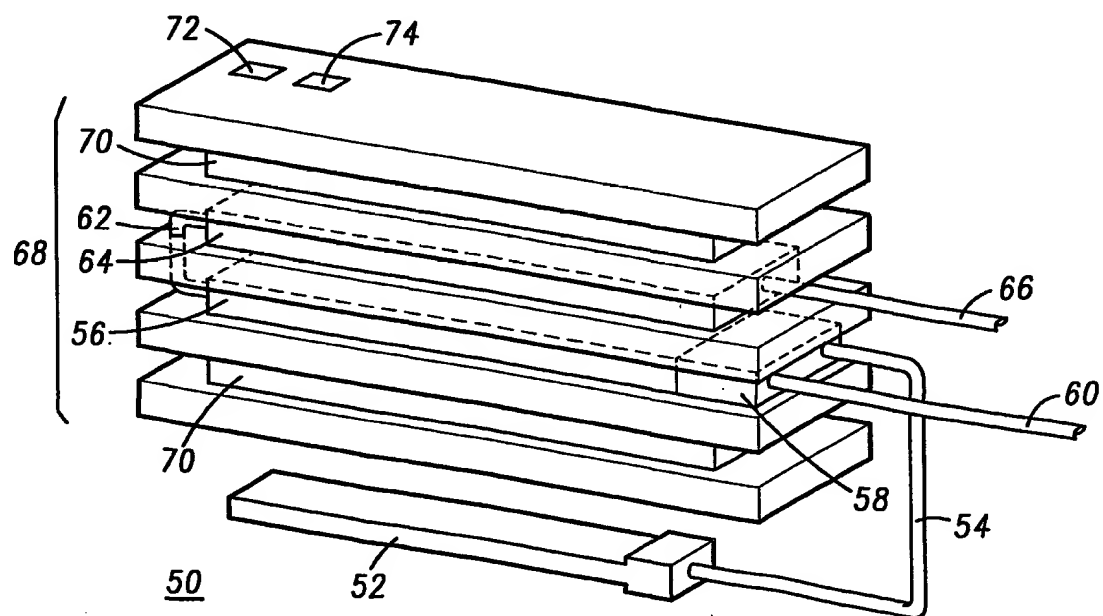
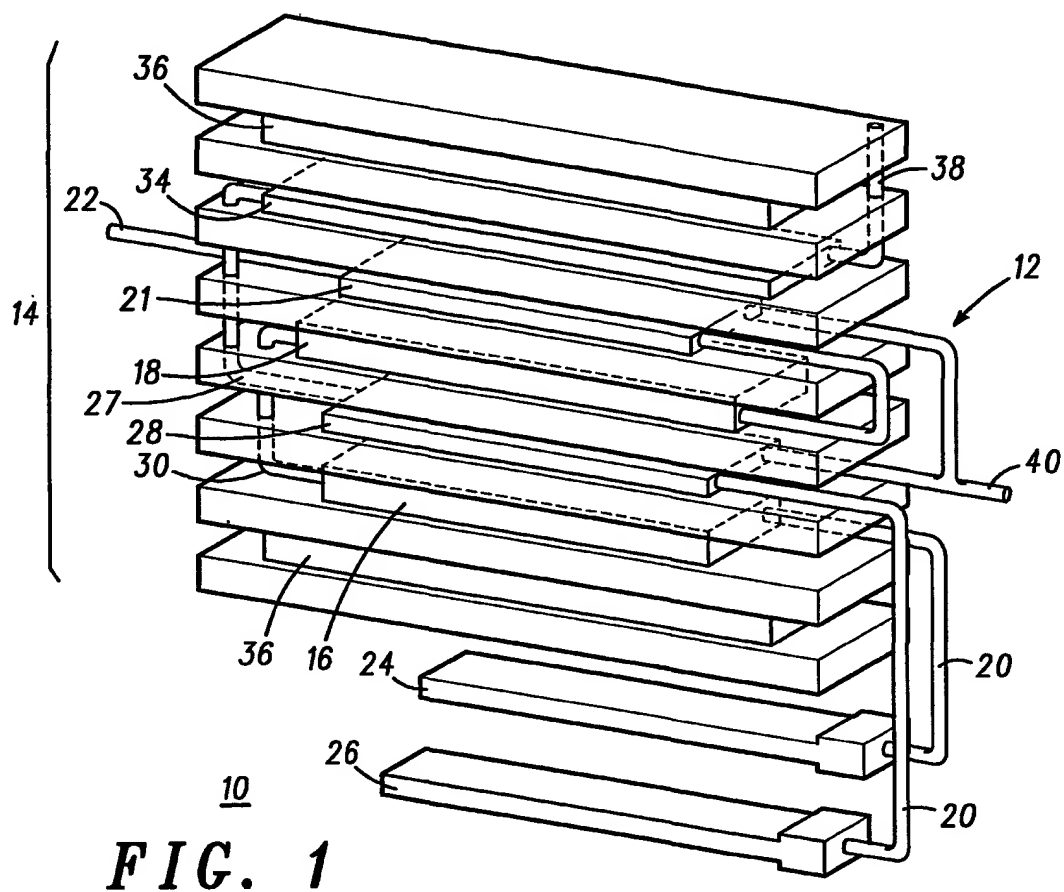
an outlet channel for hydrogen enriched gas.

12. A hydrogen generator comprising:

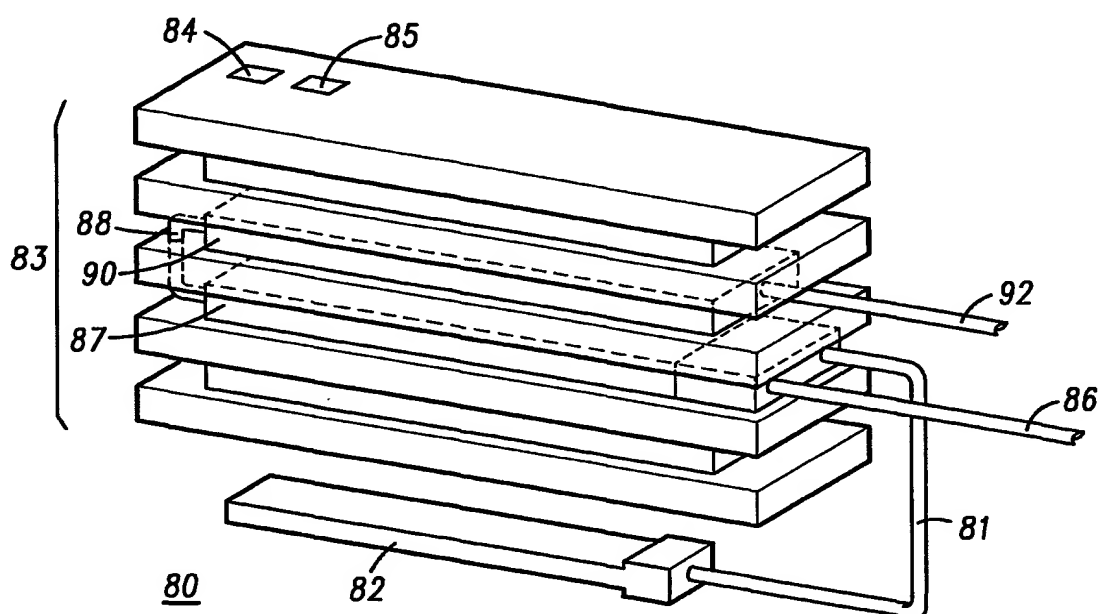
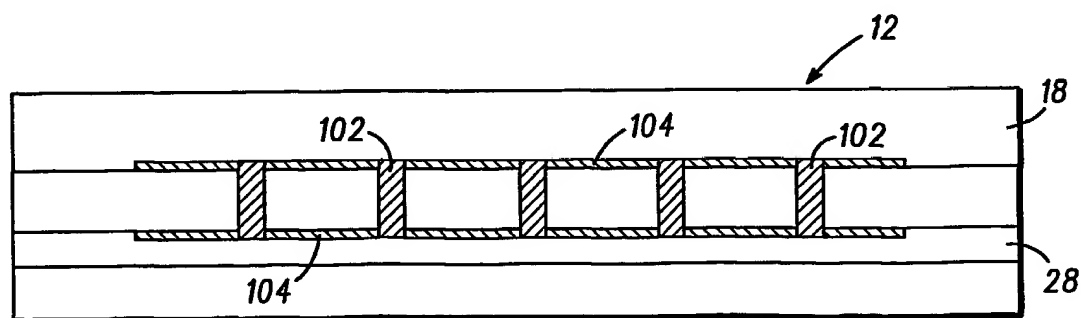
10 a three-dimensional multilayer ceramic carrier structure defining a fuel processor including a vaporization zone and a reaction zone including a reforming catalyst, at least one of the vaporization zone and the reaction zone including one of a plurality of  
15 parallel channels or at least one serpentine channel, the ceramic carrier further including an integrated heater thermally coupled to the reaction zone and the vaporization zone using thermally conductive structures;

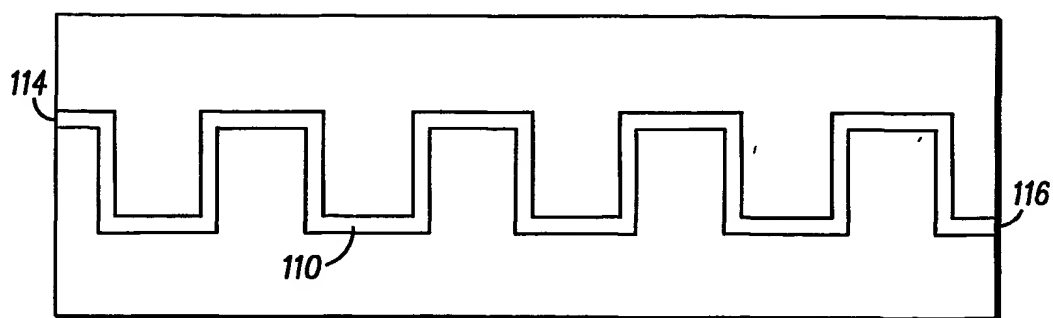
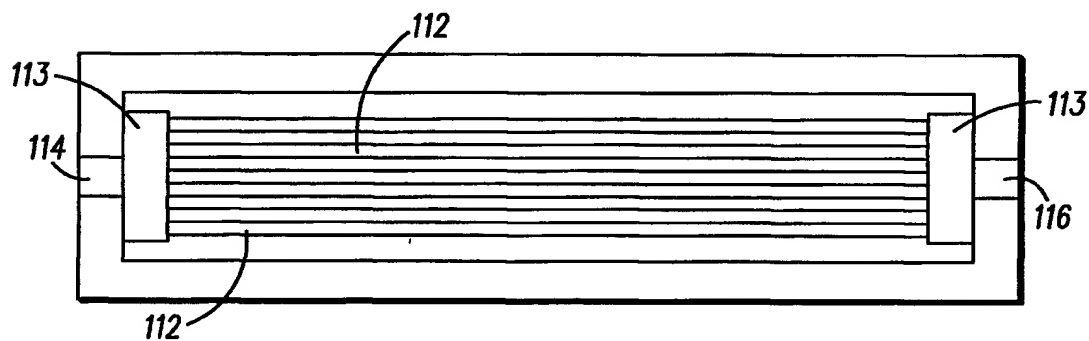
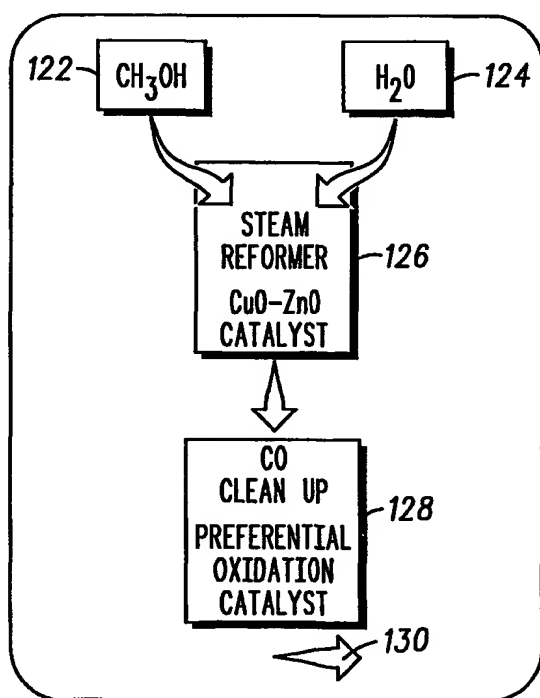
an inlet channel for liquid fuel; and

20 an outlet channel for hydrogen enriched gas.



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**FIG. 3**30**FIG. 4**

*FIG. 5**FIG. 6**FIG. 7*120